MERCURY TMDLS FOR SELECTED SEGMENTS IN THE RED RIVER AND SABINE RIVER BASINS, LOUISIANA

MERCURY TMDLS FOR SELECTED SEGMENTS IN THE RED RIVER AND SABINE RIVER BASINS, LOUISIANA

Prepared for

US EPA Region 6 Water Quality Protection Division Oversight and TMDL Team 1445 Ross Avenue, Suite 1200 Dallas, TX 75202-2733

> Contract No. 68-C-02-108 Task Order 132

> > Prepared by

FTN Associates, Ltd 3 Innwood Circle, Suite 220 Little Rock, AR 72211

FTN No. 2110-622

EXECUTIVE SUMMARY

Section 303(d) of the Federal Clean Water Act requires states to identify waterbodies that are not meeting water quality standards and to develop total maximum daily loads (TMDLs) for those waterbodies. A TMDL is the amount of a pollutant that a waterbody can assimilate without exceeding the established water quality standards for that pollutant. Through a TMDL, pollutant loads can be allocated to point sources and nonpoint sources discharging to the waterbody.

This report presents TMDLs that have been developed for mercury for several subsegments associated with lakes with mercury fish consumption advisories in the Red and Sabine River basins in Louisiana. Table ES.1 summarizes characteristics of these subsegments.

Table ES. 1. Summar	of characteristics of listed	d subsegments addressed i	in this report.
		6	

Subsegment Number	Waterbody Description	Subsegment Area (km²)	Dominant Land Use (%)
100401- 0556575	Ivan Lake	88	Forest (75.1%)
100703	Black Lake & Clear Lake	350	Forest (49.1%)
100705	Kepler Creek Lake	55	Forest (53.5%)
100709, 100709-001	Grand Bayou & Grand Bayou Reservoir	322	Forest (49.0%)
100803	Saline Bayou	139	Grassland/pasture/hay (38.6%)
101302	Iatt Lake	176	Forest (57.7%)
101501	Big Saline Bayou	124	Wetlands (61.8%)
101502	Saline Lake	157	Cultivated crops (46.2%)
101504	Saline Bayou	61	Wetlands (79.4%)
101505	Larto Lake	85	Cultivated crops (50.4%)
101506	Big Creek	312	Forest (30.4%)
110101	Toledo Bend Reservoir	3,000	Forest (41.6%)
110503	Vernon Lake	78	Forest (34.4%)

These subsegments were included on the Louisiana Department of Environmental Quality (LDEQ) final 2004 303(d) list as not supporting their fish and wildlife propagation designated uses, and were ranked as priority No. 1 for TMDL development. Atmospheric

deposition of mercury was identified as the suspected cause of impairment for the subsegments. The Mercury Action Level in Louisiana for fish consumption advisories is 0.5 mg/kg. EPA has recently promulgated a methyl mercury criterion for fish tissue of 0.3 mg/kg. There have been no known violations of the numeric mercury water quality standard in any of the listed subsegments.

The estimated mercury load to the listed subsegments included mercury atmospheric deposition from local emission sources, regional atmospheric deposition, mercury previously deposited in the watershed and transported to the water body via erosion, inflows from upstream subsegments, and point sources. Where adjacent subsegments were listed, the adjacent subsegments were bundled together when calculating the TMDLs (i.e., 100704+100709+100803, and 101501+101502+101504+101505+101506). The largest sources of mercury load to the listed subsegments were atmospheric deposition and erosion.

The wasteload allocations (WLAs) for point source contributions were set to the Louisiana mercury water quality criterion multiplied by the point source flow. The margin of safety was implicit due to conservative assumptions in the TMDL calculations. A 10% future growth component was included in the TMDLs. The TMDLs and percent reductions needed are summarized in Table ES.2.

Table ES.2. Summary of TMDLs and percent reductions.

Subsegment	TMDL (g/day)	MOS (g/day)	FG (g/day)	WLA (g/day)	LA (g/day)	Reduction
100401-0556575	3.5	implicit	0.4	0	3.1	62%
100705	5.9	implicit	0.6	0	5.3	60%
100703	22.5	implicit	2.2	0	20.3	50%
100709	22.0	implicit	2.2	0	19.8	50%
100803	9.3	implicit	0.9	0	8.4	50%
101302	3.0	implicit	0.3	0	2.7	68%
101501	1.8	implicit	0.2	0	1.6	68%
101502	8.0	implicit	0.8	0	7.2	68%
101504	1.9	implicit	0.2	0	1.7	68%
101505	5.6	implicit	0.6	0	5.0	68%
101506	11.0	implicit	1.1	0	9.9	68%
110101	59.3	implicit	5.9	0.3	53.1	51%
110503	5.6	implicit	0.6	0	5.0	17%

This TMDL report indicates that current mercury loadings to the listed subsegments are primarily from atmospheric sources. Mercury load reductions necessary to achieve the target fish tissue concentration of 0.5 mg/kg range from 17% to 68%. Consequently, significant reduction in atmospheric deposition within and outside the study areas will be necessary. A combination of ongoing and future activities under the Clean Air Act are expected to achieve reductions in atmospheric deposition of mercury that will enable reductions in fish tissue mercury concentrations.

It may be appropriate to revise these TMDLs at some point in the future based on new information gathered and analyses performed. An adaptive management approach allows the United States (US) Environmental Protection Agency (EPA) or the State to use the best information available at the time to establish the TMDL at levels necessary to implement applicable water quality standards and to make the allocations to the pollution sources. EPA recognizes that additional data and information may be necessary to validate the assumptions of the TMDL and to provide greater certainty that the TMDL will achieve the applicable water quality standards. The adaptive management approach is appropriate for these TMDLs because information on the actual contributions of mercury from both point and nonpoint sources will be much better characterized in the future. EPA expects point source loadings of mercury to be reduced primarily through mercury minimization programs developed and implemented by some point sources.

During implementation of these TMDLs, EPA expects the following activities to occur:

- 1. NPDES point source discharges will develop and implement mercury minimization plans as appropriate.
- 2. Air emissions of mercury will be reduced through implementation of the Clean Air Act regulation.
- 3. LDEQ will collect additional ambient data on mercury concentrations in water, sediment, fish, and soil.
- 4. LDEQ will develop and implement a mercury risk reduction plan that assesses all sources of mercury.

TABLE OF CONTENTS

1.0	INTR	ODUC	ΓΙΟΝ	1-1
2.0	BAC	KGROU	JND INFORMATION	2-1
	2.1	Gener	al Information	2-1
		2.1.1	Ivan Lake	2-1
		2.1.2	Kepler Creek Lake	2-1
		2.1.3	Black Lake	2-1
		2.1.4	Iatt Lake	2-2
		2.1.5	Saline Lake	2-2
		2.1.6	Toledo Bend Reservoir	2-2
		2.1.7	Vernon Lake	2-2
	2.2	Land	Use	2-3
	2.3	Water	Quality Standards and Fish Tissue Action Levels	2-3
	2.4	Point	Sources	2-4
	2.5	Nonpo	oint Sources	2-4
3.0	EXIS	TING C	CONDITIONS	3-1
	3.1	Mercu	ary in Water	3-1
	3.2	Other	Water Quality Parameters	3-1
	3.3	Fish T	Tissue Data	3-3
4.0	TMD	L DEVI	ELOPMENT	4-1
	4.1	TMDI	L Method	4-1
		4.1.1	Conceptual Framework	4-1
		4.1.2	Loading Capacity	4-4
		4.1.3	TMDL Formulation	4-4
	4.2	Requi	red Load Reductions	4-4
	4.3	Existi	ng Loads	4-4
		4.3.1	Point Sources	4-5
		4.3.2	Nonpoint Sources	4-7

TABLE OF CONTENTS (CONTINUED)

	4.4	TMDI		4-14
		4.4.1	MOS and FG	4-15
		4.4.2	WLA	4-15
		4.4.3	LA	4-15
		4.4.4	Seasonality	4-16
5.0	ONGO	OING A	ND FUTURE POLLUTANT LOADING REDUCTIONS	5-1
	5.1	Atmos	spheric mercury	5-1
		5.1.1	National and State	5-1
		5.1.2	International	5-4
	5.2	Munic	ipal dischargers	5-4
	5.3	Pollut	ion Prevention	5-5
	5.4	LDEQ	Statewide Mercury Program	5-5
6.0	PUBL	IC PAR	RTICIPATION	6-1
7.0	REFE	RENCE	ES	7-1

LIST OF APPENDICES

APPENDIX A:	Maps
-------------	------

APPENDIX B: Point Source Discharges

APPENDIX C: Water Quality Data

APPENDIX D: Fish Tissue Mercury Data

APPENDIX E: Local Mercury Air Emission point sources from 2002 National Emissions

Inventory

APPENDIX F: Sediment Mercury Data

APPENDIX G: Responses to Comments

LIST OF TABLES

Table ES. 1	Summary of characteristics of listed subsegments addressed in this report	i
Table ES.2	Summary of TMDL and percent reduction	ii
Table 1.1	Summary of 303(d) listings addressed in this TMDL Report	1-1
Table 2.1	Land use percentages for study areas	2-3
Table 2.3	Summary of NPDES point source discharges in TMDL subsegments	2-5
Table 3.1	Mercury in water data for TMDL subsegments	3-2
Table 3.2	Summary of LDEQ fish tissue mercury data associated with TMDL subsegments	3-4
Table 4.1	Mercury load reduction factors for TMDL study areas	4-5
Table 4.2	Estimated existing mercury loads to TMDL study areas	4-6
Table 4.3	Point source mercury loads	4-7
Table 4.4	Tributary mercury loads to TMDL study areas	4-8
Table 4.5	Estimated total atmospheric mercury deposition rate	4-9
Table 4.6	Local source emissions for Sabine River basin subsegments 110101, 110503	4-11
Table 4.7	Local source emissions for Red River basin subsegment 100401-0556575	4-12
Table 4.8	Local source emissions for Red River basin subsegments 100703, 100705, 100709, 100803, and 101302	4-12
Table 4.9	Local Source Emissions for Red River basin subsegments 101501, 101502, 101504, 101505, and 101506	4-13
Table 4.10	Summary of mercury TMDLs	4-14
	LIST OF FIGURES	
Figure 4.1	General mercury cycle showing atmospheric transport and deposition, point, nonpoint source and natural background contributions, and the effects of new reservoirs on mercury release into the environment	4-1
Figure 4.2	Pathways for mercury species through the aquatic ecosystem, including methylation and demethylation, evasion or loss from the water to the atmosphere, and sedimentation and burial in the sediment	4-3

1.0 INTRODUCTION

This report presents total maximum daily loads (TMDLs) for mercury for 12 Red River basin subsegments and two Sabine River basin subsegments. These subsegments were listed as impaired on the final 2004 303(d) List for Louisiana dated August 17, 2005 (Louisiana Department of Environmental Quality (LDEQ) 2005a). Table 1.1 shows the suspected sources and suspected causes for impairment in the 303(d) List, as well as the priority ranking.

Table 1.1. Summary of 303(d) listings addressed in this TMDL Report (LDEQ 2005a).

Subsegment No.	Waterbody Description	Suspected Sources	Suspected Causes	Priority Ranking (1 = highest)
100401-0556575	Ivan Lake	Atmospheric Deposition- Toxics, Source Unknown	Mercury	1
100703	Black Lake & Clear Lake	Atmospheric Deposition- Toxics, Source Unknown	Mercury	1
100705	Kepler Creek Lake	Atmospheric Deposition- Toxics, Source Unknown	Mercury	1
100709	Grand Bayou	Atmospheric Deposition- Toxics, Source Unknown	Mercury	1
100709-001	Grand Bayou Reservoir	Atmospheric Deposition- Toxics, Source Unknown	Mercury	1
100803	Saline Bayou	Atmospheric Deposition- Toxics, Source Unknown	Mercury	1
101302	Iatt Lake	Atmospheric Deposition- Toxics, Source Unknown	Mercury	1
101501	Big Saline Bayou	Atmospheric Deposition- Toxics, Source Unknown	Mercury	1
101502	Saline Lake	Atmospheric Deposition- Toxics, Source Unknown	Mercury	1
101504	Saline Bayou	Atmospheric Deposition- Toxics, Source Unknown	Mercury	1
101505	Larto Lake	Atmospheric Deposition- Toxics, Source Unknown	Mercury	1
101506	Big Creek	Atmospheric Deposition- Toxics, Source Unknown	Mercury	1
110101	Toledo Bend Reservoir	Atmospheric Deposition- Toxics, Source Unknown	Mercury	1
110503	Vernon Lake	Atmospheric Deposition- Toxics, Source Unknown	Mercury	1

The TMDLs in this report were developed in accordance with Section 303(d) of the Federal Clean Water Act and the Environmental Protection Agency's (EPA's) regulations at 40 CFR 130.7. The 303(d) Listings for other pollutants in these subsegments are being addressed by the EPA and LDEQ in other documents.

The purpose of a TMDL is to determine the pollutant loading that a waterbody can assimilate without exceeding the water quality standard for that pollutant and to establish the load reduction that is necessary to meet the standard in a waterbody. The TMDL is the sum of the wasteload allocation (WLA), the load allocation (LA), future growth (FG), and a margin of safety (MOS). The WLA is the load allocated to point sources of the pollutant of concern. The LA is the load allocated to nonpoint sources, including natural background. The MOS is a percentage of the TMDL that takes into account any lack of knowledge concerning the relationship between pollutant loadings and water quality. The FG is the portion of the TMDL that allows for future increases in loads to the waterbody.

2.0 BACKGROUND INFORMATION

A watershed–based approach was used in developing the TMDLs in this report. Therefore, those subsegments that are contiguous were considered together in developing mercury loads and targets. Overall, seven study areas were addressed in this report and are described below (Figure A.1 in Appendix A).

2.1 General Information

2.1.1 Ivan Lake

Ivan Lake (subsegment 100401-0556575) is in the Red River basin in northwest Louisiana (Figure A.2 in Appendix A). Ivan Lake is an impoundment of Caney Creek and Philips Creek located near Cotton Valley, Louisiana. Downstream of Ivan Lake, Caney Creek joins Bodcau Bayou. Ivan Lake is included in the United States Geological Survey (USGS) Hydrologic Unit 11140205. This watershed encompasses 88 km² in subsegment 100401.

2.1.2 Kepler Creek Lake

Kepler Creek Lake (subsegment 100705) is in the Red River basin in northwest Louisiana (Figure A.3 in Appendix A). Kepler Creek Lake is an impoundment of Kepler Creek, a tributary of Black Lake Bayou, with headwaters just west of Bryceland, Louisiana. Kepler Creek Lake is included in the USGS Hydrologic Unit 11140209. Subsegment 100705 encompasses approximately 55 km².

2.1.3 Black Lake

This study area includes listed tributaries to Black and Clear Lakes (subsegment 100709 – Grand Bayou), the lakes themselves (subsegment 100703), and Saline Bayou downstream of the lakes (subsegment 100803). This study area is located in the Red River basin in northwestern Louisiana (Figure A.4 in Appendix A). Subsegments 100703 and 100709 are included in the USGS Hydrologic Unit 11140209, and subsegment 100803 is included in the USGS Hydrologic Unit 11140208. This study area encompasses approximately 811 km².

2.1.4 latt Lake

Iatt Lake (subsegment 101302) is in the Red River basin in central Louisiana (Figure A.5 in Appendix A). Iatt Lake is an impoundment of Iatt Creek, which has its headwaters southwest of Winnfield, Louisiana. Iatt Lake is included in the USGS Hydrologic Unit 11140207. Subsegment 101302 encompasses approximately 176 km².

2.1.5 Saline Lake

This study area includes Saline Lake (subsegment 101502) and its tributaries (subsegment 101506 – Big Creek, 101501 – Big Saline Bayou), Saline Bayou (subsegment 101504), and Larto Lake (subsegment 101505). This study area is located in the Red River basin in east central Louisiana, near the confluence of the Red and Black Rivers (Figure A.6 in Appendix A). This study area encompasses approximately 739 km² in the USGS Hydrologic Unit 08040301.

2.1.6 Toledo Bend Reservoir

Toledo Bend Reservoir (subsegment 110101) is in the Sabine River basin along the western border of Louisiana (Figure A.7 in Appendix A). Toledo Bend Reservoir is an impoundment of the Sabine River. Subsegment 110101 encompasses approximately 3,000 km² in USGS Hydrologic Unit 12010004.

2.1.7 Vernon Lake

Vernon Lake (subsegment 110503) is in the Sabine River Basin in west central Louisiana (Figure A.8 in Appendix A). Vernon Lake is an impoundment of Bayou Anacoco, which is a tributary of the Sabine River, and is located northwest of Leesville, Louisiana.

Subsegment 110503 encompasses approximately 78 km² in USGS Hydrologic Unit 12010005.

2.2 Land Use

Land use characteristics for the study areas were compiled from the USGS 2001 National Land Cover Dataset (http://gisdata.usgs.net/website/MRLC/viewer.php). These data were based on satellite imagery from 2001 and they represent the most recent land use data available for this area. The spatial distribution of these land uses is shown on Figures A.9 through A.15 (located in Appendix A) and land use percentages are shown in Table 2.1.

Table 2.1. Land use percentages for study areas.

Study Area	Subsegments	Water	Urban	Barren	Forest	Grassland/ Pasture/Hay	Cultivated Crops	Wetlands
Ivan Lake	100401 - 0556576	1.6%	2.0%	0%	75.1%	15.8%	0%	5.6%
Kepler Creek Lake	100705	13.4%	5.0%	0%	53.5%	25.4%	0.4%	2.4%
	100703	14.8%	3.0%	0%	49.1%	19.5%	0.5%	13.2%
Black	100709	3.6%	4.6%	0%	49.0%	30.7%	0.1%	12.0%
Lake	100803	4.8%	5.6%	0%	28.2%	38.6%	8.8%	13.9%
	Total	8.2%	4.1%	0%	45.5%	27.6%	1.7%	12.8%
Iatt Lake	101302	11.5%	4.7%	0.1%	57.7%	14.5%	0.2%	11.3%
	101701	1.00/	4.40/	0.04	40.50	1.4.50		
	101501	1.8%	4.1%	0%	18.5%	11.7%	2.2%	61.8%
G 11	101502	6.1%	1.2%	0%	0.6%	1.2%	46.2%	44.7%
Saline	101504	9.4%	1.7%	0%	0.7%	0%	8.8%	79.4%
Lake	101505	13.3%	2.0%	0%	1.1%	0.1%	50.4%	33.1%
	101506	1.2%	5.7%	0%	30.4%	21.9%	20.0%	20.8%
	Total	4.4%	3.7%	0.0%	16.2%	11.5%	25.2%	39.0%
Toledo	1				l	I		
Bend Reservoir	110101	11.2%	3.6%	0.2%	41.6%	23.0%	0.1%	20.2%
Vernon Lake	110503	20.4%	13.6%	0.2%	34.4%	22.6%	0%	8.9%

2.3 Water Quality Standards and Fish Tissue Action Levels

Water quality standards for Louisiana are included in the Title 33 Environmental Regulatory Code (LDEQ 2005b). Designated uses for the subsegments addressed in this TMDL

are primary and secondary contact recreation, fish and wildlife propagation, and agriculture. The chronic numeric criterion for mercury in water to protect aquatic life in Louisiana is 0.012 µg/L.

The mercury fish consumption Action Level in Louisiana is 0.5 mg/kg (wet weight). EPA has promulgated a criterion of 0.3 mg/kg (wet weight) for methyl mercury in fish tissue.

The Louisiana water quality standards also include an antidegradation policy (LAC 33: IX.1109.A). This policy states that waters exhibiting high water quality should be maintained at that high level of water quality. If this is not possible, water quality of a level that supports designated uses of the waterbody should be maintained. Changing the designated uses of a waterbody to allow a lower level of water quality can only be achieved though a use attainability study.

2.4 Point Sources

Lists of National Pollutant Discharge Elimination System (NPDES) point source discharges in the subsegments were generated by LDEQ using their TEMPO and PTS databases. Table 2.3 is a summary of NPDES permitted dischargers in the subsegments based on these lists. A list of the discharges in the study areas is shown in Table B.1 (Appendix B). The locations of these point source discharges are shown on Figure A.16 (Appendix A). None of the NPDES discharges had permit limits for mercury. Clean sampling of municipal wastewater discharges in Arkansas found measurable mercury concentrations in the effluent of all facilities tested. Therefore, municipal wastewater discharges were considered as possible sources of mercury in these TMDLs. To be consistent with previous Louisiana mercury TMDLs, mercury loads were calculated only for municipal wastewater discharges with flow greater than 100,000 gpd (Table 2.3).

2.5 Nonpoint Sources

Atmospheric deposition is the only mercury source specified for these subsegments in the 2004 303(d) List. Significant proportions of mercury emissions are deposited locally, within 100 km of emission sources. There are approximately 46 mercury emission sources within 100 km of the subsegments included in this TMDL. However, mercury can also be transmitted

much farther, regionally or globally, before deposition. Local and regional mercury emission sources were considered in these TMDLs. In addition, mercury is often present in watershed soils, as a result of current and historical atmospheric deposition, and possibly naturally occurring, and can be transported to surface water bodies via soil erosion. Mercury also enters some of these study areas from upstream subsegments.

Table 2.3. Summary of NPDES point source discharges in TMDL subsegments.

Study Area	Number of Permitted Dischargers	Number of Municipal Wastewater Discharges with flow > 100,000 gpd
Ivan Lake Watershed (100401-0556575)	0	0
Kepler Creek Lake (100705)	7	0
Black Lake (100703, 100709, 100803)	13	1
Iatt Lake (101302)	0	0
Saline Lake (101501, 101502, 101504,		
101505, 101506)	16	0
Toledo Bend Reservoir (110101)	92	4
Vernon Lake (110503)	2	0

3.0 EXISTING CONDITIONS

3.1 Mercury in Water

Measurements of mercury in ambient water have been collected by LDEQ at water quality stations located in subsegments addressed in this TMDL report. Locations of these water quality stations are shown on Figures A.2 through A.8 (located in Appendix A). Table 3.1 shows summaries of the mercury data, including percentages of values above the mercury criterion of $0.012~\mu g$ /L. It should be noted that prior to 2002 the detection level for mercury in water was greater than the mercury water quality criterion. Starting in 2002 samples were collected and analyzed using "clean" techniques to prevent sample contamination. Results from sampling prior to 2002 are believed to reflect sample contamination rather than actual conditions in the water bodies sampled. All results from 2002 and later were less than the mercury water quality criterion. A table of data for the entire period of record at each station is included in Appendix C. These data were obtained from LDEQ.

3.2 Other Water Quality Parameters

Measurements of sulfate, total organic carbon (TOC), and pH have also been collected at these water quality monitoring stations (Appendix C). These data were obtained from LDEQ. These three constituents have been demonstrated to be correlated with fish tissue mercury concentrations, and can affect the bioavailability of mercury for methylation and subsequent uptake and bioaccumulation of methyl mercury through the food chain (Armstrong et al., 1995, EPA 1998). Water bodies with moderate sulfate (5 to 25 mg/L) and TOC (5 to 10 mg/L) concentrations provide an environment conducive to microorganisms that methylate mercury and tend to have fish with higher tissue mercury concentrations (Armstrong et al., 1995). Waterbodies with lower pH values (<5.5 su) can experience chemical mercury methylation. All of the water quality monitoring stations in the TMDL subsegments had pH values that were predominately greater than 5.5 su so chemical methylation would not be expected. Most of the sites have exhibited moderate sulfate and/or TOC concentrations (Appendix C).

Table 3.1. Mercury in water data for TMDL subsegments.

									%
	LDEQ		Period of	No. of	Minimum	Maximum	Median*	No. Values	Values
Subsegment	Station	Waterbody	Record	Values	$(\mu g/L)$	(µg /L)	(µg/L)	>0.012*	>0.012*
100703	366	Black Lake	2002, 2006	5	0.00078	0.00633	0.0045	0	0
100705	1188	Kepler Creek Lake	2002, 2006	9	0.00034	0.00303	0.00139	0	0
100709	1190	Grand Bayon	2002	4	0.00078	0.00639	0.00247	0	0
100803	1214	Saline Bayon	2002	3	0.00031	0.00536	0.00454	0	0
101302	570	Beaver Creek	1997-2000	13	<0.050	0.070	0.050	13	100
101302	1221	Iatt Lake	2002-2004	5	0.00120	0.00806	0.00351	0	0
101501	1224	Big Saline Bayou	2002, 2006	5	0.00372	0.00606	0.00548	0	0
101502	486	Johns Bayon	1993	1	<0.05	<0.05	<0.05	1	100
101504	371	Saline Bayon	2002	3	0.00059	0.00944	0.00728	0	0
101505	1225	Larto Bayou	2002	3	0.00193	0.00330	0.00255	0	0
101505	1226	Larto Lake	2002	3	0.00025	0.00623	0.00232	0	0
101506	1227	Big Creek	2002	3	0.00572	0.00832	0.00576	0	0
110503	1164	Vernon Lake	2002, 2006	4	0.00047	0.00399	0.00254	0	0
110101	1154	Toledo Bend Reservoir	2002, 2006	4	0.00033	0.00069	0.000545	0	0
* A comming detection walne for less than	on value for l	action detection							

*Assuming detection value for less than detection.

3.3 Fish Tissue Data

LDEQ has been collecting and analyzing fish samples for mercury in fish tissue since 1993. Water bodies with fish consumption advisories are sampled every two years. A number of fish sampling sites are located in the subsegments being addressed in this TMDL report. The locations of these sampling sites are shown in Figures A.2 through A.8 (Appendix A). Table 3.2 summarizes the fish tissue data that are available for the TMDL subsegments. A table of all the data is included in Appendix D.

Table 3.2. Summary of LDEQ fish tissue mercury data associated with TMDL subsegments.

Water Body	Subsegment	Fish Tissue Sites	Record	Fish Advisories	
Grand Bayou	100700 001	597	1997-2000, 2002-	Bowfin, Largemouth	
Reservoir	100709-001	587 – near Coushatta	2003, 2005	Bass	
Kepler Creek	100705	590 – N of Castor	1997, 2001, 2005	Bowfin	
Lake	100703	1188 – SE Jamestown	2003	DOWIII	
Saline Bayou	100803	3048 – near Clarence	2005	none	
Big Saline Bayou	101501	700 – E of Deville	1998, 2000		
Larto Lake	101505	711 – NE of Marksville	1988, 2001-2002	Bowfin, Largemouth	
Saline Lake	101502	376 – N of Marksville	1994	Bass, White Bass,	
		999 – SE of Deville	2000	Freshwater Drum,	
Big Creek	101506	3181 – N of Marksville	2005	Flathead Catfish,	
		1141	2001	White Crappie	
Shad Lake	101504	2892- Cross Bayou	2004		
		371 – Saline Bayou	2003		
Iatt Lake	101302	375 – NE of Colfax	1994,1999, 2004	Bowfin, Largemouth Bass	
Ivan Lake	100401- 0556575	964	1999-2002	Bowfin, Largemouth Bass	
Black Lake and Clear Lake	ke 100703	366 – N of Natchitoches	1994-1996, 1999- 2002, 2004		
		614 – Clear Lake	1997	Bowfin, Largemouth Bass, White Bass,	
		3049- Black Lake Bayou	2005	Crappie, Freshwater Drum	
		2852 – Clear Lake @ Clarence	2004		
Lake Vernon 110503		522 – S of Anacoco	1995-1997, 1999- 2001, 2004	Largemouth Bass, Flathead Catfish,	
Lake veriion	110303	1164 – NE of Standard	2002	Redear Sunfish, Bluegill Sunfish	
		374 – S of Zwolle	1994-1995, 2000		
		471 – S of Logansport	1994-1995, 1998, 2000		
		529	1995-1998, 2000-2002		
		530	1995, 2000		
			1995-1998, 2000-		
Toledo Bend		531	2001, 2005	Bowfin, Largemouth	
Reservoir	110101	532 – SW of Zwolle	1995	Bass, Freshwater	
		534	1995, 2000	Drum	
		535 – neart Toro	1995, 2000-2001	1	
		603 – SW of Logansport	1997	1	
		604 – W of Zwolle	1997	1	
		1006 – SW of Zwolle	2000		
		537	1996		

4.0 TMDL DEVELOPMENT

4.1 TMDL Method

4.1.1 Conceptual Framework

Mercury is unlike many other metals because it has a volatile phase at ambient temperatures and can be transported in a gaseous, soluble, or particulate form (Figure 4.1). Mercury is emitted to the atmosphere in both elemental gaseous Mercury(0) and divalent Mercury(II) forms. Anthropogenic direct emissions, natural emissions, and indirect re-emission of previously deposited mercury are major sources of mercury to the atmosphere (Figure 4.1). Gaseous Mercury (0) is relatively insoluble and is capable of being transported long distances and contribute to regional and global background concentrations.

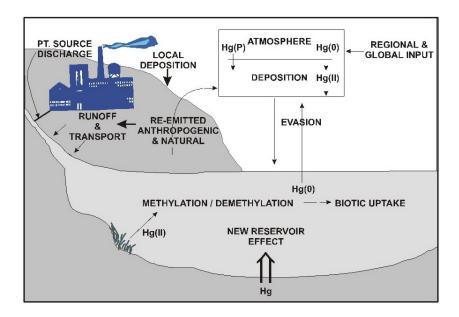


Figure 4.1. General mercury cycle showing atmospheric transport and deposition, point, nonpoint source and natural background contributions, and the effects of new reservoirs on mercury release into the environment.

Mercury(II) is much more soluble and can sorb onto particulates, so it tends to be removed from the atmosphere by both wet and dry mercury deposition closer to emission sources, within local and regional areas (EPRI 1994). Ozone or other oxidizing agents in the atmosphere can convert Mercury(0) to Mercury(1), and some Mercury(II) can also be chemically

reduced to Mercury(0). Mercury(0) can be transported long distances Local sources of deposited mercury are typically within about a 100 km radius of a site (EPA 2001). Regional sources are loosely defined as other sources within a geographical area such as the Southeast, South, or Upper Midwest, while global sources include intercontinental contributions of mercury. Atmospheric mercury deposition can include contributions from all three sources. In addition to atmospheric deposition, mercury can also enter waterbodies from point source effluent discharges and watershed nonpoint source contributions. These watershed nonpoint sources include both naturally occurring mercury (e.g., geology, soils) and atmospherically deposited mercury that can be transported to the waterbody (Figure 4.1).

The primary mercury species of concern for bioaccumulation and biomagnification through the food chain, however, are not the inorganic mercury species, but the organic form methyl mercury (Figure 4.2). Inorganic mercury deposited in waterbodies can be converted to methyl mercury. Sulfate reducing bacteria are thought to be the agent responsible for the majority of methyl mercury production in aquatic systems (Beyers et al., 1999, Compeau and Bartha 1987, Gilmour and Henry 1991), and in situ production is often a significant source of methyl mercury in aquatic systems (Benoit et al., 1998, Gilmour et al., 1998, Mason et al., 1999).

Methyl mercury binds with protein in muscle tissue of fish and other living organisms. Methyl mercury is lost very slowly from fish tissue, on the order of years (Trudel and Rasmussen 1997). Therefore, methyl mercury concentrations continue to biomagnify or increase in concentration throughout the life of the fish as long as methyl mercury is in the environment and in its prey species. Older, larger fish typically have higher mercury concentrations than younger, smaller fish. Several factors can affect the availability of inorganic mercury for conversion to methyl mercury. If sulfides or dissolved organic matter are present, they can bind inorganic mercury so that it is not available for conversion to methyl mercury (Benoit et al., 1999; Ravichandran 2004). Inorganic mercury can also join with more complex polysulfides or other chemicals and become easier for methylating bacteria to use (Benoit et al., 1999, 2000, 2001). In addition, recent research indicates that inorganic mercury tends to become less likely to be converted to methyl mercury the longer it is in a waterbody (Hintelmann et al., 2002); more recently deposited inorganic mercury is more reactive.

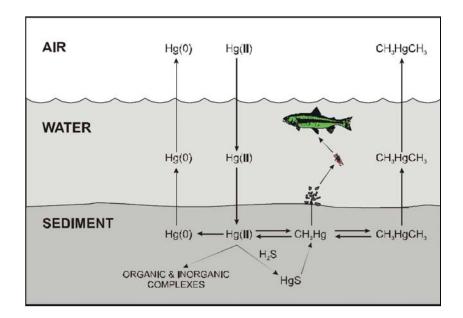


Figure 4.2. Pathways for mercury species through the aquatic ecosystem, including methylation and demethylation, evasion or loss from the water to the atmosphere, and sedimentation and burial in the sediment (after Winfrey and Rudd 1990).

Methylating microorganisms, such as sulfur reducing bacteria, live in anaerobic (zero dissolved oxygen) environments in the sediments of wetlands, streams, rivers, and lakes or reservoirs. New reservoirs (i.e., less than 15 to 20 years old) create environments that are particularly suitable for methylating bacteria so fish tissue mercury concentrations in new reservoirs are typically higher than fish tissue mercury concentrations in older reservoirs.

In summary, TMDLs for mercury must consider that mercury can exist as a gas as well as in solution or particulate forms. Mercury loads arise from atmospheric deposition contributed by both local and regional/global emission sources, point source effluent discharges, natural geological formations, and soils. However, after deposition or loading to the system, it can also be lost through volatilization and re-enter the atmospheric pool. It is the organic form as methyl mercury that is biologically accumulated and magnified through the food chain. Once in fish, it is lost very slowly and continues to accumulate through time.

4.1.2 Loading Capacity

The loading capacity of waterbodies differ based on a site specific basis due to (1) inputs or load of mercury to the waterbody, (2) environmental conditions within the waterbody that mediate methylation and bioaccumulation, and (3) the food web or food chain through which mercury bioaccumulates (Armstrong et al., 1995).

4.1.3 TMDL Formulation

A three-step approach was used to estimate loading capacity and the reductions required to achieve the designated fishable use in the TMDL study areas. In the first step, required load reductions were estimated based on existing and target fish tissue mercury concentrations. In the second step, mercury loading to the study areas was estimated. In the third step, the TMDLs were estimated by applying the estimated required load reductions to the estimated existing mercury loads to the study areas.

4.2 Required Load Reductions

The target for the TMDLs in this report is the Louisiana fish consumption action level (0.5 mg/kg). For each TMDL study area the average tissue concentration for the fish species with the highest value was used to calculate the mercury load reduction factor. For those study areas with more than one listed water body with fish tissue mercury data, the highest average value from all of the water bodies was used. In most cases bowfin was the fish species with the highest average tissue mercury concentration. The load reduction factor was calculated by dividing the target fish tissue concentration by the average measured fish tissue concentration. This number is essentially the portion of the existing load that would be the target load or one minus the percent reduction. Table 4.1 shows the data used and the resulting load reduction factors along with the equivalent percent reduction for each TMDL study area.

4.3 Existing Loads

The existing mercury load to the study areas was assumed to consist of loads from both point and nonpoint sources. Point sources were NPDES permitted municipal wastewater

treatment plants either with mercury permit limits or flow greater than 100,000 gpd. Nonpoint sources load included tributary inputs, atmospheric deposition inputs from local and regional/global emission sources, and watershed soil erosion inputs. Estimated loads from these sources are summarized in Table 4.2. The methods used to estimate these loads are described below.

Maximum average fish tissue mercury concentration Reduction **Percent** Study Area (mg/kg) Fish species factor reduction 0.38 Bowfin Ivan Lake 1.3 62% Kepler Creek Lake 1.24 Bowfin 0.40 60% Black Lake 1.01 **Bowfin** 0.49 50% Iatt Lake 1.53 Bowfin 0.32 68% Saline Lake 1.61 **Bowfin** 0.31 68% Toledo Bend 1.03 Bowfin 0.48 51% Reservoir Bluegill Vernon Lake 0.60 0.83 17% Sunfish

Table 4.1. Mercury load reduction factors for TMDL study areas.

4.3.1 Point Sources

None of the NPDES permitted discharges identified in the study areas were found to have a mercury permit limit. However, because measurable mercury levels have been found in discharge from municipal wastewater treatment plants, point source mercury loads were calculated for all NPDES discharges identified as municipal wastewater treatment plants (including LAG56 and LAG57 permits) with an expected or design flow of greater than 100,000 gpd. EPA believes it is appropriate to assume that these discharges contain mercury levels equal to $0.012~\mu g/L$. Information for the discharges included in the TMDLs (Table 4.3) was obtained by FTN Associates, Ltd. from LDEQ's Electronic Data Management System. Facility mercury load was estimated by multiplying expected or design flow (whichever was available) by $0.012~\mu g/L$. Table 4.3 shows the estimated point source mercury loads.

Table 4.2. Estimated existing mercury loads to TMDL study areas.

					Local	Local Source	Regional Source	Source			
	Point I	Point Source	Tribu	Tributary	Atmospheric Deposition	pheric	Atmospheric Denosition	pheric	Soil	Soil	Total
Study Area (subsegments)	g/yr	%	g/yr	%	g/yr	%	g/yr	%	g/yr	%	g/yr
Ivan Lake	0	0	0	0		0.03	51	1	3,316	86	3,368
Kepler Creek Lake (100705)	0	0	66	2	ю	0.05	191	С	5,166	95	5,429
Black Lake (100703, 100709, 100803)	0	0	1,778	4	42	1	2,377	9	35,861	68	40,060
latt Lake (101302)	0	0	777	22	11	0.3	615	18	2,061	59	3,464
Saline Lake (101501, 101502, 101504, 101505, 101506)	0	0	0	0	115	0.3	5,433	16	27,684	83	33,232
Toledo Bend Reservoir (110101)	94	0.21	923	2	727	2	980,6	20	34,246	92	45,076
Vernon Lake (110503)	0	0	529	22	24	1	306	12	1,575	99	2,435

Mercury Mercury Concentration Study **Facility NPDES** Flow Load **Mercury Load** Name Area No. **MGD** (lb/day) $(\mu g/L)$ (g/yr) Town of 40×10^{-5} LA0033308 0.4 0.012 66.31 Logansport Town of 8×10^{-5} LA0020354 0.8 0.012 13.26 Zwolle Toledo Village of Bend 1×10^{-5} Pleasant LA0038946 0.1 0.012 1.66 Reservoir Hill Town of LA0056502 8×10^{-5} 0.75 0.012 12.43 Many 59 x 10⁻⁵ **Total** 93.66

Table 4.3. Point source mercury loads.

4.3.2 Nonpoint Sources

4.3.2.1 Tributary Inputs

Several of the listed subsegments receive inflow from upstream subsegments. These inflows have mercury loads associated with them that contribute to the mercury load in the listed subsegments. These loads were estimated using mercury data from the LDEQ water quality monitoring stations associated with the upstream subsegments and recorded flows at USGS gaging stations. To estimate outflow from the upstream subsegments, an average flow per unit area was calculated for each of the USGS gaging stations and multiplied by the area within the subsegment. The average annual flow for the entire period of record at the gaging stations was used to calculate the flow per unit area. A number of the upstream subsegments did not contain USGS gaging stations, so nearby stations were used to estimate outflow from the subsegment. Table 4.4 shows the upstream subsegments associated with the TMDL study areas and the data used to estimate the tributary loads to the TMDL study areas.

Table 4.4. Tributary mercury loads to TMDL study areas.

		Upstream		Average		Average	Tributary
	Upstream	Area	ΜÓ	Mercury		gage flow	Load
Study Area	Subsegments	(mi²)	Station	$(\mu { m g/L})^1$	USGS Gage	(cfs/mi²)	(g/yr)
Ivan Lake	None						0
Kepler Creek Lake (100705)	100704	26.3	283	0.00366	7352000 Saline Bayou near Lucky	1.148	9.86
Black Lake	100702	433.1	1187	0.003926	7349000 Bayou Dorcheat near Minden	1.032	1567
100709, 100803)	100802	107.8	1205	0.002544	7353000 Saline Bayou near Clarence	0.862	210.9
latt Lake (101302)	101303	170.4	1222	0.004504	7353500 Natachie Creek near Montgomery	1.134	777.3
Saline Lake (101501, 101502, 101504, 101505, 101506)	None	1	1	1		1	0-
Toledo Bend Reservoir (110101)	HUC 12010002 in Texas	2769.7	1154²	0.000528 ²	8022040 Sabine River near Texas	0.707	922.9
Vernon Lake	110501	47.9	1162	0.005012	802800 Bayou Anacoco near	1.328	284.7
(110503)	110502	60.7	1163	0.003398	Rosepine		244.6

Average of values measured with clean sampling and analysis techniques, beginning in 2002.
 No mercury measurements with detection levels less than 0.05 mg/L were found for Sabine River upstream of Toledo Bend River, so reservoir measurements were used.

4.3.2.2 Regional Atmospheric Deposition

Data for atmospheric deposition of mercury was obtained from the National Atmospheric Deposition Program (NADP) website. There are NADP mercury deposition monitoring stations reasonably close to the TMDL study areas (for a map showing locations of all of the NADP monitoring sites, see http://napd.sws.uiuc.edu/mdn/sites.asp). Data from monitoring stations TX21 and LA23 were used to represent atmospheric deposition of mercury in the study areas. Data were available from both of these stations for 2001 through 2005. The average value of the wet deposition at these sites for this period was $11.7 \,\mu\text{g/m}^2/\text{yr}$ (Table 4.5). An estimate of the total atmospheric deposition was based on the assumption that dry deposition is about 50% to 60% of wet deposition (Auwarter 2000) resulting in a total atmospheric deposition of $18.7 \,\mu\text{g/m}^2/\text{yr}$. Wet deposition is the mercury removed from the atmosphere during rain events. Dry deposition is the mercury removed from the atmosphere on dust particles, sorption to vegetation, gaseous uptake by plants, or other input during non-rainfall periods (EPA 1997).

Table 4.5. Estimated total atmospheric mercury deposition rate.

NA DD GC 41	•	Mercury Deposition
NADP Station	Year	$(\mu g/m^2/yr)$
TX21	2001	15.0
TX21	2002	8.6
TX21	2003	9.2
TX21	2004	12.5
TX21	2005	7.6
LA23	2001	14.5
LA23	2002	12.3
LA23	2003	11.6
LA23	2004	17.4
LA23	2005	8.3
	Average =	11.7
	$Dry + Wet = Average \times 1.6 =$	18.7

The total direct atmospheric deposition mercury load to the listed water bodies was calculated by multiplying the total atmospheric deposition rate (18.7 μ g/m²/yr) by the sum of the wetland and water land uses for the study areas (see Table 2.2) The part of the atmospheric

deposition load to the TMDL study areas coming from regional or global emissions sources was estimated by subtracting the local emissions load from the total atmospheric deposition load (Table 4.2).

4.3.2.3 Local Emissions Atmospheric Deposition

The data from the TX21 and LA23 deposition monitoring stations includes both local emission sources similar to those in Texas and Louisiana, and regional/global input. Local atmospheric deposition for the TMDL study areas was estimated based on data from the 2002 National Emissions Inventory (NEI). The NEI is a complete national inventory of stationary and mobile sources that emit hazardous air pollutants (HAPs). County summaries of NEI point source emissions data from 2002 were downloaded from the NEI web site (www.epa.gov/ttn/chief/net/2002inventory.html).

In this TMDL, local sources are defined as sources within the study areas and within a distance of 100 km around the study areas boundary. The area within which these local sources are located is referred to as the "airshed". The NEI reports sources listed by county; therefore the airshed boundary is determined by county boundaries and if a portion of a county falls within 100 km of the study area, then the entire county is included as part of the airshed. Several of the study areas were close enough together that their airsheds were the same, so only four airsheds were defined. The airshed boundaries for the study areas are shown in Figures A.17 through A.20 (Appendix A). The mercury emissions for each source found within these airsheds are included in Appendix E.

The NEI reports emissions of total mercury. As discussed in Section 4.1.1 Mercury(II) is the form that is most likely to be removed by wet and dry deposition closer to emission sources (i.e., within 100 km). Therefore we want to use just the Mercury(II) emissions when estimating atmospheric deposition of mercury from local emissions. A number of studies have been done characterizing mercury emissions from a variety of sources and the portion of those emissions that occur as Mercury(II). Tables 4.6 through 4.9 show the Mercury(II) emissions for each source category that contributes to the local atmospheric deposition for each TMDL study area, calculated from the NEI data using Mercury(II) percentages from EPA (2005a). The total

mercury emissions load for the airshed was converted to an areal load by dividing by the area of the airshed. The local emissions direct atmospheric deposition mercury load to the listed water bodies (Table 4.2) was calculated by multiplying the areal load by the sum of the wetland and water land uses for the study area (see Table 2.2).

The distance from the emission source, the forms of the mercury in the emissions, other pollutants in the emissions and the atmosphere, and the weather patterns of precipitation and prevailing wind are important factors in determining where mercury released to the air will deposit.

Table 4.6. Local source emissions for Sabine River basin subsegments 110101, 110503.

Source Category	Total Mercury Emissions (tons/yr)	% Particulate Mercury(II)	% Gaseous Mercury(II)	Mercury(II) Emissions (tons/yr)
Electricity Generation	0.0022	20	30	0.0011
Industrial Boilers	0.026	20	30	0.013
Chlor-Alkali Production	0.61	0	5	0.030
Steel Manufacture	0.063	10	10	0.013
Secondary Metal Production	0.28	10	10	0.056
Paper Production (Kraft Pulping)	0.0085	20	30	0.0042
Miscellaneous Manufacturing	0.055	20	30	0.027
Total		•		0.14

Table 4.7. Local source emissions for Red River basin subsegment 100401-0556575.

Source Category	Total Mercury Emissions (tons/yr)	% Particulate Mercury(II)	% Gaseous Mercury(II)	Mercury(II) Emissions (tons/yr)
Electricity Generation	0.0012	20	30	0.00062
Industrial Boilers	0.0048	20	30	0.0024
Steel Manufacture	0.063	10	10	0.013
Paper and Wood Production	0.00009	10	30	0.000045
Miscellaneous Manufacturing	0.046	20	30	0.023
Total				0.039

Table 4.8. Local Source emissions for Red River basin subsegments 100703, 100705, 100709, 100803, and 101302.

Source Category	Total Mercury Emissions (tons/yr)	% Particulate Mercury(II)	% Gaseous Mercury(II)	Mercury(II) Emissions (tons/yr)
Electricity Generation	0.0014	20	30	0.00068
Industrial Boilers	0.011	20	30	0.0053
Paper and Wood Production	0.0051	20	30	0.0025
Miscellaneous Manufacturing	0.048	20	30	0.024
Total				0.033

Table 4.9. Local Source Emissions for Red River basin subsegments 101501, 101502, 101504, 101505, and 101506.

Source Category	Total Mercury Emissions (tons/yr)	% Particulate Mercury(II)	% Gaseous Mercury(II)	Mercury(II) Emissions (tons/yr)
Electricity Generation	0.0014	20	30	0.00068
Industrial Boilers	0.010	20	30	0.0052
Chlor-alkali Production	0.46	0	5	0.023
Chemical Manufacturing	0.003	10	10	0.0006
Plywood Production	0.0016	10	10	0.00032
Sawmill Operations	0.0005	20	30	0.00025
Miscellaneous Manufacturing	0.0024	20	30	0.0012
Total				0.031

4.3.2.4 Mercury Load Associated with Soil Erosion

The mercury loads for the study areas associated with transport of eroded material into the water bodies was calculated using literature erosion rates for forest, pasture, and cropland land uses. The land use areas for the study areas were based on USGS 2001 National Land Cover Dataset (http://gisdata.usgs.net/website/MRCL/viewer.php) data as presented in Section 2.2. The erosion rates for pasture and cropland were set to average erosion rates reported for these land uses for Louisiana in the 1997 National Resources Inventory (NRI); these values were 0.2 tons/acre/year for pasture and 3.3 tons/acre/year for cropland. The NRI was conducted and published by the US Department of Agriculture (USDA) National Resources Conservation Service (USDA 2000). Forest erosion rates were not available for the study area parishes in the NRI, therefore the forest erosion rate was set 0.2 tons/acre/year based on information from other sources (Bloodworth and Berc 1981, Novotny and Chesters 1981, USDA Forest Service 1999).

Erosion rates for barren land were not available, so barren land was assumed to have an erosion rate that is similar to cropland. The resulting estimates of tons of sediment per year transported to the water bodies were multiplied by average sediment mercury concentrations

measured in the subsegments (Appendix F) to estimate study area mercury loads associated with soil erosion (Table 4.2).

4.4 TMDL

The total allowable mercury loads for the study areas (i.e., the TMDLs) were calculated based on the existing load, assuming a linear relationship between mercury loads to the waterbodies and mercury concentrations in fish tissue. In other words, it was assumed here that reducing the mercury loads to the waterbodies by a factor of 2 (for example) would eventually result in a reduction of mercury concentrations in fish tissue by a factor of 2. The assumption of this linear relationship between mercury load and fish tissue mercury concentration is consistent with steady-state assumptions and the use of bioaccumulation factors, and has been demonstrated in field experiments in the Florida Everglades (Atkeson et al., 2003) and Canada (Orihel et al., 2006). Based on this assumption, the TMDLs were calculated as the existing mercury loads multiplied by the reduction factors (Section 4.2). The TMDL components and load reductions are summarized in Table 4.10.

Table 4.10. Summary of mercury TMDLs.

Subsegment	TMDL (g/day)	MOS (g/day)	FG (g/day)	WLA (g/day)	LA (g/day)
100401-0556575	3.5	implicit	0.4	0	3.1
100705	5.9	implicit	0.6	0	5.3
100703	22.5	implicit	2.2	0	20.3
100709	22.0	implicit	2.2	0	19.8
100803	9.3	implicit	0.9	0	8.4
101302	3.0	implicit	0.3	0	2.7
101501	1.8	implicit	0.2	0	1.6
101502	8.0	implicit	0.8	0	7.2
101504	1.9	implicit	0.2	0	1.7
101505	5.6	implicit	0.6	0	5.0
101506	11.0	implicit	1.1	0	9.9
110101	59.3	implicit	5.9	0.3	53.1
110503	5.5	implicit	0.5	0	5.0

4.4.1 MOS and FG

The MOS accounts for any lack of knowledge or uncertainty concerning the relationship between LAs and water quality. In this case, it accounts for uncertainty and variability related to fish tissue mercury concentrations, estimates of loading, and assumption of a linear relationship between watershed mercury load and fish tissue mercury concentration. The MOS for these TMDLs is implicit due to the following conservative assumptions made in calculating the TMDLs:

- 1. Calculations of mercury load associated with soil erosion assume no loss of mercury from any mechanism during transport.
- 2. Mercury loading to the water bodies is considered 100% available for uptake.
- 3. For municipal wastewater treatment plants with flows greater than 100,000 gpd, it was assumed that $0.012 \mu g/L$ of mercury was discharged from each facility, however, actual discharge of mercury from these facilities may be less.

An additional 10% of the TMDL was set aside to account for uncertainty associated with FG.

4.4.2 WLA

Point sources of mercury were not numerous in the listed subsegments, and accounted for significantly less than 1% of the mercury loads. Therefore, point source loads were not reduced in these TMDLs. The WLA for point source contributions was set to the design flow multiplied by the mercury water quality criterion (0.012 μ g/L).

4.4.3 LA

The LA for nonpoint sources was set to the TMDL minus the FG and the WLA. Since tributary mercury concentrations are already below the Louisiana mercury criterion, no changes were anticipated to the tributary loads. The reduction in the nonpoint source mercury loads would result from reductions in the atmospheric deposition and sediment loads.

4.4.4 Seasonality

Wet deposition is greatest in the winter and spring seasons. Mercury loads fluctuate based on the amount and distribution of rainfall, and variability of localized and regional/global sources. While an average daily load is established here, the average annual load is of greatest significance because mercury bioaccumulates over the life of the fish and the resulting risk to human health from fish consumption is a long-term phenomenon. Thus, daily or weekly inputs are less meaningful than total annual loads over many years. The use of annual loads allows for integration of short-term and seasonal variability. Inputs should continue to be estimated through wet deposition and additional monitoring.

Mercury methylation is expected to be highest during the summer. High temperatures promote biological activity and reservoirs are stratified with anoxic hypolimnions. Based on the enhanced methylation and higher predator feeding rates during this period, mercury bioaccumulation is expected to be greatest during the summer. However, given the long depuration times for fish and relatively mild winters in Louisiana, seasonal changes in fish tissue mercury body burden are expected to be relatively small. Inherent variability of mercury concentrations between individual fish of the same and/or different size categories is expected to be greater than seasonal variability.

5.0 ONGOING AND FUTURE POLLUTANT LOADING REDUCTIONS

Table 4.2 shows that existing mercury loadings to the project study areas are primarily from nonpoint sources. As discussed in Section 4.2, mercury load to the project study areas will need to be reduced 17% to 68% to achieve the TMDL target of 0.5 mg/kg mercury in fish tissue.

5.1 Atmospheric mercury

There is good evidence that reducing atmospheric deposition loads of mercury can reduce fish tissue mercury concentrations. Results from the METAALICUS project suggest that fish tissue concentrations are most responsive to changes in mercury loads entering waterbodies through direct deposition to the water surface (compared to changes in mercury deposition to the watershed that may be transported to the waterbody) (Blanchfield et al., 2005). Reduction of mercury emissions within Florida is believed to be the cause of a more than 60% decline in mercury concentrations in Everglades's fish (Atkeson et al., 2003). The EPA study of the benefits of the Clean Air Mercury Rule suggests that the reduction of mercury deposition resulting from the Rule would result, on average, in about a 6% reduction in fish tissue mercury concentrations in Louisiana by 2020 (EPA 2005b). Because the majority of the mercury load to the study areas is from erosion of previously deposited mercury and direct atmospheric deposition, the fish mercury concentrations may take decades to decline in response to decreased mercury emissions and deposition (Chen et al., 2005).

5.1.1 National and State

In 1997, EPA reported that approximately 60% of the atmospheric mercury deposited in the US was emitted from US sources (EPA 1997). Facilities in Louisiana are subject to both state (LAC 33: III. Chapter 51) and federal mercury air emission rules. As rules and standards pursuant to the Clean Air Act have been developed, proposed, and promulgated since 1990, compliance by emitting sources as well as actions taken voluntarily have already begun to reduce emissions of mercury to the air across the US (www.epa.gov/air/mercuryrule/charts.html). The

EPA expects a combination of ongoing activities will continue to reduce mercury emissions to the air over the next decade.

The EPA currently regulates emissions of mercury and other HAPs under the maximum achievable control technology ("MACT") program of Section 112 of the Clean Air Act, and under a corresponding new source performance standard ("NSPS") program under Sections 111 and 129 of the Act. Section 112 authorizes the EPA to address categories of major sources of HAPs, including mercury, by issuing emissions standards that, for new sources, are at least as stringent as the emissions control achieved by the best performing similar source in the category, and for existing sources, are at least as stringent as the average of the best performing top 12% (or five facilities – whichever is greater) of similar sources. The EPA may also apply these standards to smaller area sources, or choose to apply less stringent standards based on generally available control technologies ("GACT"). Sections 111 and 129 direct the EPA to establish MACT-equivalent standards for each category of new and existing solid waste incineration units, regulating several specified air pollutants, including mercury.

Based on the EPA's National Toxics Inventory, the highest emitters of mercury to the air include coal-burning electric utilities, municipal waste combustors, medical waste incinerators, mercury cell chlor-alkali plants, and hazardous waste combustors. The EPA has issued a number of regulations under Sections 111, 112, and 129 to reduce mercury pollution from several of these source categories. Relevant regulations that the EPA has established to date under the Clean Air Act include those listed below.

- 1. Coal-burning electric utilities accounted for the greatest percentage of US mercury air emissions in 1990. In 1999 they accounted for over 40% of the US mercury air emissions. In March 2005, the EPA issued the Clean Air Intestate Rule and the Clean Air Mercury Rule. When fully implemented these rules will reduce mercury emissions from coal-burning electric utilities by nearly 70% from 1999 emissions levels.
- 2. Medical waste incinerators (MWIs) accounted for about 24% of US mercury air emissions in 1990. The EPA issued emission standards under Sections 111 and 129 for MWIs on August 15, 1997. The implementation deadline for these standards was September 2002. This rule reduced mercury emissions from MWIs by about 97% from 1990 emission levels.

- 3. The source category of municipal waste combustion (MWC) accounted for about 20% of US mercury air emissions in 1990. The EPA issued final regulations under Sections 111 and 129 for large MWCs on October 31, 1995. Large combustors or incinerators were required to be in compliance with the rule by December 2000. These regulations reduce mercury emissions from these facilities by about 90% from 1990 emission levels.
- 4. Mercury cell chlor-alkali plants accounted for about 4.5% of US mercury air emissions in 1994 to 1995. In December 2003, the EPA issued mercury emission standards for these facilities under Section 112. When fully implemented, these standards will reduce mercury emissions from mercury cell chlor-alkali plants by about 50%.
- 5. Hazardous waste combustors (HWCs) accounted for about 2.5% of US mercury air emissions in 1990. In February 1999, the EPA issued emission standards under Section 112 for these facilities, which include incinerators, cement kilns, and lightweight aggregate kilns that burn hazardous waste. This regulation has not been implemented, pending resolution of a lawsuit. This regulation is expected to reduce mercury emissions from HWCs by more than 50% from 1990 emission levels.

These promulgated regulations, when fully implemented and considered together with actions discussed below that will reduce the mercury content of waste, are expected to reduce national mercury emissions caused by human activities by about 50% from 1990 levels.

There are also several national programs for reducing mercury emissions from the waste stream. In 1996 the US eliminated the use of mercury in most batteries under the Mercury Containing and Rechargeable Battery Management Act. In 2006 EPA initiated the National Vehicle Mercury Switch Recovery Program, a program to reduce mercury emissions by up to 75 tons over the next 15 years by removing mercury-containing light switches from scrap vehicles before they are recycled into steel. In addition, voluntary measures to reduce use of mercury containing products, such as the voluntary measures committed to by the American Hospital Association, also will contribute to reduced emissions from waste combustion.

It is possible that the cumulative effect of additional standards and voluntary actions will reduce mercury emissions from human activities in the US by more than 50% from 1990 levels. In 1999, mercury emissions had already dropped 45% from 1990 levels. Mercury deposition

modeling of the influence of the Clean Air Interstate Rule suggests that mercury deposition in the Louisiana study areas would be reduced less than $5\mu g/m^2$ by 2020 (EPA 2005b).

5.1.2 International

Mercury emitted to the air can travel the globe and be deposited outside national boundaries, contributing to mercury issues in other countries. The United Nations Environment Programme established its Mercury Programme in 2003. This program has the long term objective "to substantially reduce or eliminate uses and anthropogenic releases of mercury through the implementation of national, regional and global actions, thereby significantly reducing global adverse impacts on health and the environment" (UNEP 2006). Through this program, a number of global partnerships for mercury reduction have been initiated, dealing with global sources such as chlor-alkali plants, products, artisanal and small scale gold mining, and coal fired utilities. In addition, a global partnership for research into mercury fate and transport has also been initiated under this program. The US participates in these global partnerships.

The US is also a member of the Commission of Environmental Cooperation (CEC), with Canada and Mexico, under the North American Agreement on Environmental Cooperation. The CEC has developed the North American Regional Action Plan on Mercury. This plan has the goal of reducing anthropogenic mercury emissions through international and national initiatives, and has provisions regarding waste management; risk management approaches to address mercury emissions, processes, operation and products; and research, monitoring, modeling, inventory, and communication activities.

5.2 Municipal dischargers

These TMDLs focus on those facilities likely to be discharging mercury. EPA expects LDEQ to systematically identify any dischargers that are significant sources of mercury. EPA will work with LDEQ to establish mechanisms for demonstrating that the WLAs in these TMDLs are met.

If a facility is found to discharge mercury at levels above $0.012 \,\mu\text{g/L}$, a mercury minimization plan may be required. EPA expects that the State of Louisiana, as the duly

authorized permitting authority, will determine any additional necessary elements of a mercury characterization/minimization plan, considering the size and nature of the affected facility. Local characteristics such as water velocity, bed substrate, oxygen content, and microbial community structure all contribute to methylation potential. Since these characteristics have not been defined for each of the discharges in each subsegment, there exists the potential that effluent containing mercury may cause localized exceedances of the criteria and therefore, minimization plans and/or numeric limits may be necessary to assure that the discharge does not cause and/or contribute to an exceedance of the applicable water quality standards. In conclusion, due to uncertainty in the TMDL analysis, mercury minimization plans and/or numeric limits may be necessary to assure compliance with the water quality standards. Through these actions, over long-term, it can be demonstrated that WLAs are being met.

5.3 Pollution Prevention

Source reduction, through product and innovation, is the key element to pollution prevention. The US industrial demand for mercury dropped 75% from 1988 to 1997 (http://www.epa.gov/mercury). Reductions in mercury use are driven by voluntary efforts and by increasingly strict federal and state regulations, such as increasing regulation of mercury in products or outright bans on the use of mercury in products for which alternatives are available. For example, in 1996 EPA eliminated the use of mercury in most batteries under the Mercury Containing and Rechargeable Battery Management Act. Other voluntary measures such as the commitment by the American Hospital Association to reduce the use of mercury-containing products will continue to decrease the amount of mercury available in the waste stream. Next to source reduction, recycling is fundamental to mercury pollution prevention. When mercury must be used and recycling is not a possibility, proper disposal is critical to reducing the potential of dispersion to the environment.

5.4 LDEQ Statewide Mercury Program

The LDEQ has identified mercury as one of its priorities and is addressing mercury risk through a statewide mercury initiative. It is the intent of LDEQ to assess all sources of mercury to the environment in the state and to develop strategies to reduce public health risks associated with mercury. A series of public meetings were held with participation from various industry sectors and non-governmental organizations. In addition, meetings on risk communication have been, and continue to be, conducted for the purpose of enhancing public awareness relative to mercury and mercury exposure.

The approach of this initiative is intended to be exhaustive and comprehensive, looking at all sources of mercury with consideration given to methods of controlling releases to the environment. Potential action items include pollution prevention strategies, waste minimization, non-essential mercury-containing device phase-outs, recycling enhancements through rule development (such as Universal Waste Rule), remediation of known sites of mercury contamination, comprehensive approaches to locating and remediating legacy sites, rule development to minimize permitted mercury emissions and discharges, and enhanced public outreach to educate the public on efforts that can be conducted locally and within the home to enjoin the mercury reduction initiative. This approach, used in the Louisiana Mercury Risk Reduction Plan, will result in the greatest environmental benefit when applied on a regional and national scale. The LDEQ and EPA will continue to develop this statewide mercury reduction strategy to its fullest potential, promoting and supporting its use in adjacent states and regions.

LDEQ continues its aggressive commitment to implementing a comprehensive statewide mercury program. The following excerpts from the recent LDEQ publication *Resource Guide to Understanding Mercury in Louisiana's Environment: 2003 Mercury Report* highlight some of the management strategies that will advance attainment of the reduction goals defined by these TMDLs (LDEQ 2003).

- Design and construction regulations for landfills to help ensure that mercury-laden materials do not leak from them.
- Historically, electrical switches in some natural gas meters contained mercury. Spills from these meters contaminated the ground and became sources of mercury to the environment. Since 1991, several natural gas pipeline companies, with oversight from LDEQ, voluntarily cleaned the mercury from the environment around contaminated natural gas meter sites. To date, approximately 5,000 sites have been checked for mercury contamination and 2,500 that were contaminated have been cleaned.

 Recycling played a large part in not only reducing the amount on mercury used by industries, but also reducing the amount released to the environment. LDEQ's Recycling Section maintains a current list of all recyclers in the state, sorted by commodity.

Over the past 5 years LDEQ has worked to expand its statewide mercury monitoring program. The primary objective of this program was to determine statewide mercury contamination levels of fish commonly eaten in Louisiana, as well as mercury concentrations in sediments, water, and epiphytic plant material, and mercury loadings from atmospheric deposition.

Continued fish tissue data collection provides input for analyses of risks to human health due to consumption of mercury-contaminated fish. This allows Louisiana Department of Health and Hospitals (LDHH) and LDEQ to address public concerns regarding the safety of fish consumption from many waterbodies. Epiphytic plant material is used to help further define the significance of atmospheric sources of mercury. Results of the epiphytic plant material analyses, together with fish tissue, water and sediment concentration information, will continue to help address questions regarding sources of mercury. Additional local and statewide remedial actions can be more effectively targeted to reduce mercury sources by combining data generated from this and previous projects and the knowledge of LDEQ field personnel. This project will also provide baseline data that can be used for ongoing trend analysis.

LDEQ's sampling site selection continues to evolve and is based on several needs. New sits are sampled to expand the number of waterbodies tested. Recently, sites were selected in basin subsegments in which no previous sampling has occurred. Currently, nearly all waterbodies with fish populations sufficient to support human health risk assessment inputs have been sampled for mercury contamination. Waterbodies are resampled if LDHH determines additional samples are needed to make a decision regarding fish consumption advisories.

Beginning in October 1998, LDEQ implemented an air monitoring program designed to assess the geographical extent and quantity of atmospheric mercury deposition. Air monitors currently exist at the Southeastern University Campus in Hammond, Louisiana; McNeese State University in Lake Charles, Louisiana; at the Louisiana State University in Chase, Louisiana;

and in Alexandria, Louisiana in Rapides Parish. Samples are tested for wet deposition of total mercury during rainfall events. If rainfall occurs samples are collected weekly. LDEQ's air monitoring sites are part of the NADP Mercury Deposition Network. Weekly data from October 1998 through present are available. The data show mercury levels are being regularly detected in rainwater. The data are analyzed by the NADP. Any interested party may access the data at the following website: http://nadp.sws.uiuc.edu/mdn.

LDEQ adheres to well-defined sampling procedures when collecting mercury data. This program is an important tool for LDEQ in evaluating the progress of the mercury reductions prescribed by these TMDLs. LDEQ's targeted data collection efforts in subsegments with fish consumption advisories will provide the data necessary to ultimately remove the fish consumption advisory or revise the TMDL at some point in the future, if warranted.

6.0 PUBLIC PARTICIPATION

When EPA establishes a TMDL, federal regulations require EPA to publish a public notice and seek comment concerning the TMDL. The TMDLs in this report were prepared under contract to EPA. After these TMDLs were developed, EPA prepared a notice seeking comments, information, and data from the general and affected public. Comments, and additional information were submitted during the public comment period, and these TMDLs were revised accordingly. Responses to submitted comments and information are included as Appendix G. EPA has transmitted the final TMDLs to LDEQ for implementation and for incorporation into LDEQ's current water quality management plan.

7.0 REFERENCES

- Armstrong, M.A, P.Burge, S. Evans, J. Giese, T. McChesney, J. Nix, A. Price, K. Thornton, D. Turman. 1995. Arkansas Mercury Task Force. Report Submitted to the Governor, State of Arkansas. AR Dept. of Pollution Control and Ecology. Little Rock, AR.
- Atkeson, T., D. Axelrod, C. Pollman, G. Keeler. 2003. Integrating Atmospheric Mercury Deposition and Aquatic Cycling in the Florida Everglades: An Approach for Conducting a Total Maximum Daily Load Analysis for an Atmospherically Derived Pollutant. Florida Department of Environmental Protection. Tallahassee, FL.
- Auwarter, A.G. 2000. Savannah River Mercury TMDL Data Report. US Environmental Protection Agency, Region 4. Athens, GA.
- Benoit, J.M., C.C. Gilmour, R.P. Mason, G.F. Riedel. 1998. Behavior of mercury in the Patuxent River estuary. Biogeochemistry. 40: 249-265.
- Benoit, J.M., C.C. Gilmour and R.P. Mason. 2000. The influence of sulfide on solid phase mercury bioavailability for methylation by pure cultures of Desulfobulbous proprionicus (1 PR 3). Submitted to Env. Sci. Technol.
- Benoit, J.M., C.C. Gilmour, and R.P. Mason. 2001. The influence of sulfide on solid-phase mercury bioavalibility for methylation by pure cultures of *Desulfobulbus propionicus* (1pr3). Environmental Science and Technology. 35(1): 127-132.
- Benoit, J.M., C.C. Gilmour, R.P. Mason, and A. Heyes. 1999. Sulfide controls on mercury speciation and bioavailability to methylating bacteria in sediment pore water. Env. Sci. Technol. 33: 951-957.
- Beyers, D.W., J.A. Rice, and W.H. Clements. 1999. Evaluating biological significance of chemical exposure to fish using a bioenergetics-based stressor-response model. Canadian Journal of Fisheries and Aquatic Sciences. 56: 823-829.
- Blanchfield, P.J. Rudd, R. Harris, H. Hintelmann. 2005. The response of Fish Mercury Levels to Increased Inorganic Mercury Loading: The METAALICUS Study. Poster presented at Eighth Annual Conference on Mercury as a Global Pollutant. August 6-11, 2006, Madison, WI.
- Bloodworth, H. and J.L. Berc. 1998. Cropland acreage soil erosion and installation of conservation buffer strips: Preliminary estimates from the 1997 National Resource Inventory. USDA-NRCS. Washington, DC.
- Chen, C.W., J.W. Herr, R.A. Goldstein. 2005. Science-based Regional Mercury TMDL Assessment. Poster presentation at the 8th International Conference on Mercury as a Global Pollutant, August 6-11 2005, Madison, WI.
- Compeau, G.C. and R. Bartha. 1987. Effect of salinity on mercury-methylating activity of sulfate reducing bacteria in estuarine sediments. Applied and Environmental Microbiology, 53: 261-265.

- EPA. 1997. Mercury Study Report to Congress. EPA-452/R-97-003. Office of Air Quality Planning & Standards and Office of Research and Development. US. Environmental Protection Agency. Washington, DC.
- EPA. 1998. South Florida Ecosystem Assessment: Monitoring for Ecosystem Restoration. Final Technical Report Phase I EPA 904-R-98-002. Region 4 Science and Ecosystem Support Division and Office of Research and Development. Athens, GA.
- EPA. 2001. Total Maximum Daily Load (TMDL) for Total Mercury in Fish Tissue Residue in the Middle and Lower Savannah River Watershed. US Environmental Protection Agency, Region 4. Athens, GA.
- EPA. 2005a. Emissions Inventory and Emissions Processing for the Clean Air Mercury Rule (CAMR). US Environmental Protection Agency. Research Triangle Park NC.
- EPA. 2005b. Regulatory Impact Analysis of the Final Clean Air Mercury Rule. EPA-452/R-05-003. US Environmental Protection Agency. Research Triangle Park, NC.
- EPRI. 1994. Mercury Atmospheric Processes: A synthesis Report. Electric Power Research Institute. EPRI/TR-104214. Palo Alto, CA.
- Gilmour, C.C. and E.A. Henry. 1991. Mercury methylation in aquatic systems affected by acid deposition. Environmental Pollution. 71: 131-169.
- Gilmour, C.C., G.S. Riedel, M.C. Ederlington, J.T. Bell, J.A. Benoit, G.A. Gill, M.C. Stordal. 1998. Methyl mercury concentrations and production rates across a trophic gradient in the northern Everglades. Biogeochemistry. 40: 327-345.
- Hintelmann, H., R. Harris, A. Heyes, J. Hurley, C. Kelly, D. Krabbenhoft, S. Lindberg, J.W.M. Rudd, K. Scott, V. St. Louis. 2002. Reactivity and mobility of new and old mercury deposition in a boreal forest ecosystem during the first year of the METAALICUS study. Environmental Science and Technology. 36: 5034-5040.
- LDEQ. 2003. Resource Guide to Understanding Mercury in Louisiana's Environment: 2003 Mercury Report. Louisiana Department of Environmental Quality. Baton Rouge, LA
- LDEQ. 2005a. Louisiana 2004 Final Intergrated Report, Appendix A. Online at www.deq.Louisiana.gov/portal/Portals/0/planning/305b/2004/04IR1-DRAFT-Appendix A with Final U.S. EPA Additions-August 17, 2005.pdf.
- LDEQ. 2005b. Title 33 Environmental Quality, Part IX Water Quality. Online at www.deq.Louisiana.gov/portal/Portals/0/planning/regs/title33/33v09.doc.
- Mason, R.P., N.M. Lawson, A.L. Lawrence, J.J. Linear, J.G. Lee, G.R. Sheu. 1999. Mercury in the Chesapeake Bay. Marine Chemistry. 65: 77-96.
- NADP. 2006. National Atmospheric Deposition Program/Mercury Deposition Network, NADP/MDN Monitoring Locations TX 21 and LA23. http://nadp.sws.uiuc.edu/nadpdata/mdnsites.asp.
- Novotny, V., G. Chesters. 1981. Handbook of Nonpoint Pollution Sources and Management. Van Nostraut Reinhold Company. New York, NY.

- Orihel, O.M., M.J. Paterson, C.C. Gilmour, R.A. Bodaly, P.J. Blanchfield, H. Hintelmann, R.C. Harris, J.W.M. Rudd. 2006. Effect of loading rate on the fate of mercury in littoral mesocosms. Environmental Science and Technology 40(19):5992-6000.
- Ravichandran, M. 2004. Interactions between mercury and dissolved organic matter a review. Chemosphere. 55: 319-331.
- Trudel, M. and J.B. Rasmussen. 1997. Modeling the elimination of mercury in fish. Env. Sci. Technol. 31: 1716-1722.
- UNEP (United Nations Environment Programme). 2006. UNEP Chemicals Mercury Programme. United Nations Environment Programme. Geneva, Switzerland.
- USDA Forest Service. 1999. Ozark-Ouachita Highland Assessment: Vol. 3-Aquatic Conditions. USDA Forest Service. Southern Research Station. Asheville, NC.
- US Department of Agriculture. 2000. Summary Report: 1997 National Resources Inventory (revised December 2000). Natural Resources Conservation Service, Washington DC; and statistical Laboratory, Iowa State University, Ames, IA